

Determination of the Upper Critical Field of a Single Crystal LiFeAs: The Magnetic Torque Study up to 35 Tesla

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We report on the upper critical field B_{c2} of a superconducting LiFeAs single crystal with $T_c \sim 16$ K, determined from magnetic torque measurements in dc-magnetic fields up to 35 T and at temperatures down to 0.3 K. B_{c2} at 0.3 K is obtained to be 26.4 T and 15.5 T for the applied field $B_a \parallel ab$ and $B_a \parallel c$, respectively. The anisotropy parameter $\Gamma = B_{c2}^{ab}/B_{c2}^c$ is ~ 3 at T_c and decreases to 1.7 as $T \rightarrow 0$, showing rather isotropic superconductivity. While B_{c2} is orbitally-limited for $B_a \parallel c$, the spin-paramagnetic effect is evident in the temperature dependence of B_{c2} for $B_a \parallel ab$.

KEYWORDS: iron-based superconductor, LiFeAs, upper critical field, anisotropy

Since the discovery of superconductivity in LaFeAs(O,F) with $T_c = 26$ K,¹⁾ a variety of related compounds containing FeAs-layers has been found to exhibit superconductivity.²⁾ The parent compounds $R\text{FeAsO}$ (R =rare earth, “1111” system) and $A\text{Fe}_2\text{As}_2$ (A =alkaline earth or Eu, “122” system) with the ZrCuSiAs - and ThCr_2Si_2 -type structures, respectively, undergo antiferromagnetic and structural transitions. The transitions can be suppressed by several kinds of doping effects^{1,3-6)} or application of pressure,⁷⁻⁹⁾ and T_c reaches ~ 56 K in some compounds.¹⁰⁻¹²⁾ The magnetic long range order usually competes with superconductivity, but the fluctuation likely plays a crucial role in the pairing mechanism of the Fe-based high- T_c systems. It is also of interest that, around the optimal condition where T_c shows its maximum, deviation from conventional Fermi-liquid behavior has been observed such as $\rho \sim T$,^{13,14)} anomalous Hall angle,¹³⁾ an enhancement of effective masses¹⁵⁾ etc. These superconducting and normal-state features bear resemblance with those widely reported for strongly-correlated electron systems including cuprates and heavy fermion compounds.

The title compound LiFeAs, categorized into the “111” system with CeFeSi -type structure, has distinctive characteristics: (i) the stoichiometric superconductivity with T_c as high as ~ 17 K,¹⁶⁾ (ii) no experimental evidence for the magnetic/structural transitions,^{17,18)} and (iii) single crystals with high quality (residual resistivity ratio up to 50).^{19,20)} Therefore, LiFeAs provides a unique opportunity to probe the intrinsic properties of the Fe-based high- T_c superconductivity.

Up to now, there are few reports on systematic measurements of the upper critical field B_{c2} of Fe-based superconductors to address the issue of the pair-breaking mechanism. This is mainly due to the fact that high-

T_c superconductors including Fe-based systems generally have extremely high B_{c2} . In many cases, accordingly, the low-temperature behavior is extrapolated from the high temperature data around T_c , which may lead to misleading conclusions. A precise determination of B_{c2} over a whole temperature range could provide important clues to the pair-breaking mechanism of high- T_c superconductivity.

Here, we present the first report on the whole temperature dependence of B_{c2} and its angular variation of a LiFeAs single crystal, using a magnetic torque technique with a 35 T dc resistive magnet.

Single crystals LiFeAs were prepared from high-purity constituent elements in a ratio of $\text{Li}:\text{Fe}:\text{As} = 2:1:2$ by a self-flux method.^{20,21)} The residual resistivity ratio is as high as 45, showing the high quality of the crystals. Temperature dependence of the dc magnetic susceptibility of LiFeAs was measured with an applied field B_a of 1 mT for $B_a \parallel ab$, in a Magnetic Property Measurement System (MPMS: Quantum Design) around T_c . As displayed in the main panel of Fig. 1, the bulk superconductivity was confirmed from the clear diamagnetic transition below $T_c \sim 16$ K with approximately 16 % (97 %) of the superconducting Meissner (shielding) volume fraction for (zero-)field-cooled process. A tiny piece taken from the same crystal was mounted on a piezoresistive microcantilever with a small amount of grease (see, inset photo of Fig. 1), attached to a rotator probe, and inserted into a ^3He cryostat (Heliox: Oxford). The angle θ is the angle between the applied field B_a and the c axis: $\theta = 0^\circ$ for $B_a \parallel c$ and $\theta = 90^\circ$ for $B_a \parallel ab$. The magnetic torque was measured with a water-cooled dc resistive magnet in fields up to 35 T and at temperatures down to 0.3 K. Since the magnetic torque, proportional to $M \times B_a$, is very small at $\theta = 0^\circ$, the torque is measured at $\theta = 5^\circ$ to improve the signal-noise ratio, which is specified as

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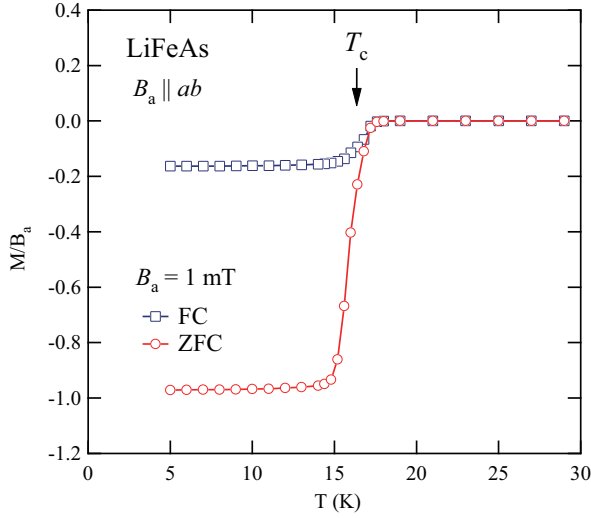


Fig. 1. (Color online) Magnetization divided by applied magnetic field, M/B_a , vs T of a single crystal LiFeAs around the transition temperature, T_c , in a field of $B_a = 1$ mT applied parallel to the ab plane. Results measured in zero-field-cooled (ZFC) and field-cooled (FC) processes are displayed. A tiny piece cleaved from the same crystal was mounted on a microcantilever for the magnetic torque measurement as shown in the inset photograph. The field angle θ is the angle between the c axis and the applied magnetic field B_a .

$B_a \parallel c$. We confirmed that the difference of B_{c2} between the two angles is negligible from the angular dependence of B_{c2} .

Figure 2 shows the field dependence of the torque signals of LiFeAs up to 35 T at fixed temperatures down to 0.3 K for (a) $B_a \parallel ab$ and (b) $B_a \parallel c$. One can see highly hysteretic behavior between the field-up and -down sweeps, which is more significant at low temperatures.²² The hysteresis of the torque response, the irreversible curve, appears in the superconducting mixed state when the pinning force is strong enough to trap the flux lines. In this study, we define the upper critical field B_{c2} as the field where the irreversibility disappears.²³ To accurately determine B_{c2} by minimizing a drift of the torque signals, we made the small-loop measurements around B_{c2} at each temperature, as shown by the dashed curves in (a) and (b) for $T = 0.3$ K, in addition to the large-loop ones between $B_a = 0$ and above B_{c2} . The definition of B_{c2} is illustrated in the inset of Fig. 2(a), where the vertical axis represents the difference of torque signals between field-up and -down sweeps of a small loop. Note that the shapes of the small loops near B_{c2} for $B_a \parallel ab$ and $B_a \parallel c$ are quite different. It could be possible that the pinning mechanism is different for the two field orientations, although the detail remains to be clarified.

Figure 3 displays thus determined B_{c2} of LiFeAs as a function of T for $B_a \parallel ab$ and $B_a \parallel c$. The B_{c2} curves are consistent with the results obtained by specific heat (C), ac-susceptibility (χ_{ac}), and ^{75}As nuclear magnetic resonance (NMR) measurements using samples of the same batch,²⁴ and are qualitatively similar to other results.^{25,26} As indicated by the dashed curves, the data can be well fitted using a Werthamer-

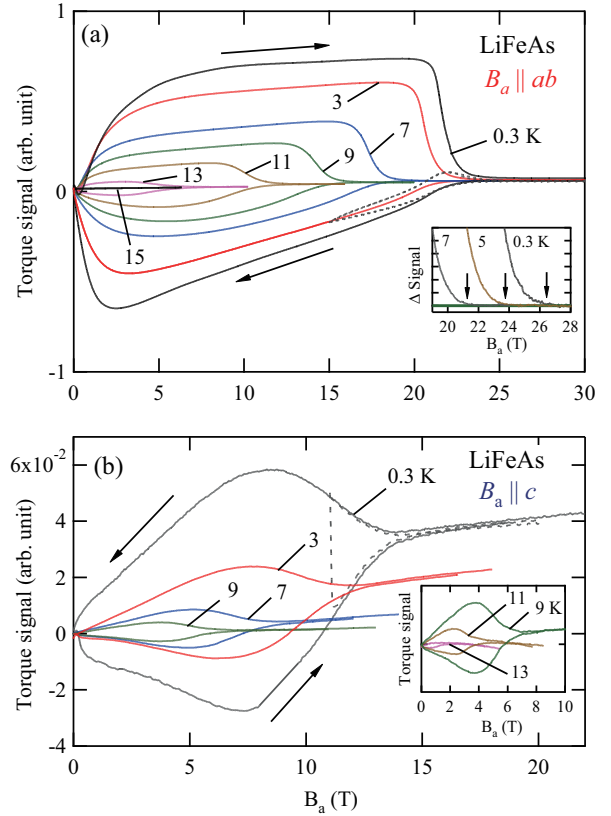


Fig. 2. (Color online) Magnetic torque signals of LiFeAs as a function of B_a at various temperatures down to 0.3 K for (a) $B_a \parallel ab$ and (b) $B_a \parallel c$ (inset: magnified view). The solid curves represent the data obtained in field-up and -down sweeps between $B_a = 0$ and above B_{c2} , where the arrows indicate the sweep directions. Small-loop measurements around B_{c2} , as shown by the dashed curves at $T = 0.3$ K, were performed at each temperature. In this study, B_{c2} was defined as the field where the difference between field-up and -down signals in a small loop becomes zero within experimental error as illustrated in the inset of (a).

Helfand-Hohenberg (WHH) formula containing the spin-paramagnetic and orbital pair-breaking effects.²⁷ The fits give the Maki parameter $\alpha = 2.30$ and 0.75 for $B_a \parallel ab$ and $B_a \parallel c$, respectively, where $T_c = 15.5$ K is fixed.²⁸ Because the torque signal, $\propto M \times B_a$, diminishes at low fields, it is difficult to unambiguously determine the initial slope $dB_{c2}/dT|_{T=T_c}$. Instead, the values of $dB_{c2}/dT|_{T=T_c}$ are estimated to be 4.43 and 1.44 T/K for $B_a \parallel ab$ and $B_a \parallel c$, respectively, from the relation $\alpha = -0.52 dB_{c2}/dT|_{T=T_c}$. These values are comparable to those obtained from ac- χ .²⁴ The orbital critical fields B_{c2}^* at $T = 0$ are estimated to be 47.2 and 15.3 T for $B_a \parallel ab$ and $B_a \parallel c$, respectively, from a dirty limit formula $B_{c2}^*(0) = 0.69 T_c dH_{c2}/dT|_{T=T_c}$.²⁷ The Pauli-Clogston paramagnetic limit $B_{po} = 1.84 T_c$ ²⁹ is 28.5 T. The Ginzburg-Landau (GL) coherence length ξ is obtained to be $\xi_{ab} = 4.64$ nm and $\xi_c = 1.50$ nm, using $B_{c2}^{*ab}(0) = \Phi_0/2\pi\xi_{ab}(0)\xi_c(0)$ and $B_{c2}^{*c}(0) = \Phi_0/2\pi\xi_{ab}(0)^2$, where $\Phi_0 = 2\pi\hbar/2e = 2.07 \times 10^{-15}$ T m² is the flux quantum. At $T = T_c$, the anisotropy of the effective masses, m_{ab}^*/m_c^* , is about 0.11 , using $B_{c2}^{*ab}/B_{c2}^{*c} = (m_c^*/m_{ab}^*)^{0.5}$. These superconducting parameters are summarized in

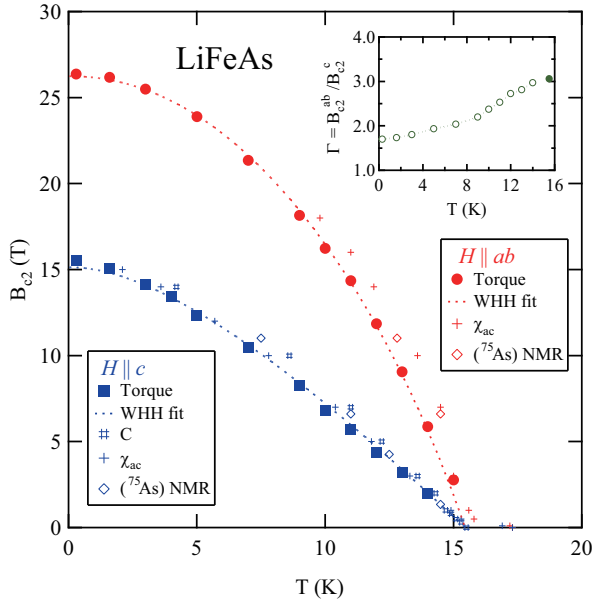


Fig. 3. (Color online) B_{c2} vs T of LiFeAs for $B_a \parallel ab$ and $B_a \parallel c$. The dashed curves indicate fits to the data based on the WHH theory (see text). For comparison, data deduced from C , χ_{ac} , and ^{75}As -NMR measurements are shown.²⁴⁾ The inset shows the T -dependence of the anisotropy parameter $\Gamma = B_{c2}^{ab}/B_{c2}^c$. The data at T_c (●) is obtained from the WHH fit.

Table I. Superconducting parameters of LiFeAs and KFe_2As_2 ,³⁰⁾ for comparison, obtained from the WHH fits; α : Maki parameter, λ_{so} : Spin-orbit scattering parameter, B_{c2}^* : Orbital critical field, B_{po} : Paramagnetic critical field, ξ : GL coherence length.

	LiFeAs		KFe_2As_2 ³⁰⁾	
	$B \parallel ab$	$B \parallel c$	$B \parallel ab$	$B \parallel c$
α	2.30	0.75	2.30	0.340
λ_{so}	0.51	∞	0.36	∞
T_c (K)	15.5		2.79	
$-\frac{dB_{c2}}{dT} _{T_c}$ (T/K)	4.43	1.44	3.8	0.71
$B_{c2}(0.3\text{ K})$ (T)	26.4	15.5	4.40	1.25
$B_{c2}^*(0)$ (T)	47.2	15.3	8.44	1.25
B_{po} (T)	28.5		5.13	
ξ (nm)	4.64	1.50	16.3	2.45

the Table I, together with those of a stoichiometric superconductor KFe_2As_2 ,³⁰⁾ for comparison.

It is interesting to note that $B_{c2}^{*c}(0) < B_{po} < B_{c2}^{*ab}(0)$. This relation results in the orbitally limited B_{c2} for $B_a \parallel c$, as indicated by $\lambda_{so} = \infty$, and strongly spin-paramagnetically limited B_{c2} for $B_a \parallel ab$, as indicated by that $B_{c2}^{ab}(0.3\text{ K}) \ll B_{c2}^{*ab}(0)$, and effectively reduces the B_{c2} anisotropy at low temperatures. A similar trend, namely the trend that the weaker orbital effect for $B_a \parallel ab$ is partly compensated for by the spin-paramagnetic effect to yield a reduced anisotropy, is clearly seen in a stoichiometric superconductor KFe_2As_2 with low T_c of 2.8 K³⁰⁾ (see, Table I), as well as in the high- T_c systems including $(\text{Ba},\text{K})\text{Fe}_2\text{As}_2$,^{31,32)} $\text{Ba}(\text{Fe},\text{Co})_2\text{As}_2$,^{33,34)} and “11”-type $\text{Fe}(\text{Se},\text{Te})$.^{35–38)}

Figure 4 shows (a) the torque signals vs B_a at 0.3 K for several angles, θ , and (b) B_{c2} vs θ at

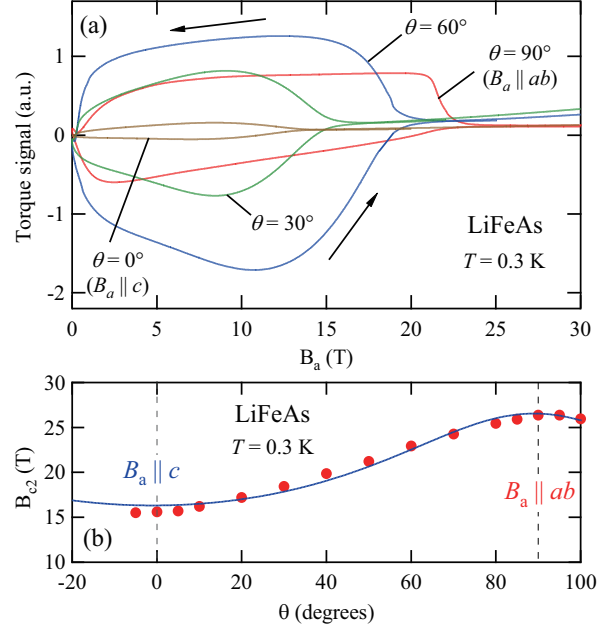


Fig. 4. (Color online) (a) Torque signals of LiFeAs as a function of B_a at 0.3 K for several angles θ , where $\theta = 0$ and 90° correspond to $B_a \parallel c$ and $B_a \parallel ab$, respectively. (b) θ -dependence of B_{c2} determined at $T = 0.3\text{ K}$. The solid curve is a fit by a GL formula (see text).

0.3 K. The solid curve in Fig. 4(b) is a fit to the $B_{c2}(\theta)$ data using a Ginzburg-Landau (GL) formula,³⁹⁾ $B_{c2}(\theta) = B_{c2}^c / (\cos^2 \theta + \Gamma^{-2} \sin^2 \theta)^{0.5}$. The fit yields $B_{c2}^c = 16.3\text{ T}$ and $\Gamma = 1.63$. Note that the GL formula is based on the orbital effect whereas the spin-paramagnetic effect is crucial for $B_a \parallel ab$ in the present case. The quality of the fit is therefore rather limited as indicated by the deviations of the data points from the fit curve. The inset of Fig. 3 shows the T -dependence of the anisotropy parameter Γ , defined as $\Gamma = B_{c2}^{ab}/B_{c2}^c$. As temperature decreases, Γ decreases from $\Gamma = 3.1$ at T_c (from the WHH fit). At 0.3 K, $B_{c2}^{ab} = 26.4\text{ T}$ and $B_{c2}^c = 15.5\text{ T}$, which give $\Gamma = 1.7$. The low-temperature variation of Γ is similar to those observed in the “122” and “11” systems,^{31–38)} but the value of Γ in LiFeAs is slightly larger. In the “122” and “11” systems, $\Gamma = 2 \sim 3$ at T_c and Γ appears to approach ~ 1 at 0 K.^{31–38)} This is markedly different from the results reported for the “1111” system. In $\text{NdFeAs}(\text{O},\text{F})$, for example, Γ is 9.2 at T_c ⁴⁰⁾ (no report for Γ as $T \rightarrow 0$ because of the large B_{c2}). The larger value of Γ in the “1111” system can be ascribed to the more two-dimensional Fermi surface structures than those in the “111”, “122” and “11” systems, as expected from band structure calculations.⁴¹⁾ Similarly, the slightly larger Γ of LiFeAs than those of the “122” and “11” systems might be due to the more two-dimensionality of LiFeAs.⁴¹⁾ In KFe_2As_2 , however, there is an exceptionally large anisotropy ($\Gamma = 6.8$ at T_c , see Table I) for the “122” system,³⁰⁾ which is, on the other hand, in accord with the large resistivity anisotropy $\rho_c/\rho_{ab} \sim 40$.³⁰⁾ In contrast, ρ_c/ρ_{ab} is reported to be as small as 3.3 in LiFeAs, and $2 \sim 5$ in $\text{Ba}(\text{Fe},\text{Co})_2\text{As}_2$.⁴²⁾

To conclude, we have performed high-field magnetic

torque measurements of a LiFeAs single crystal up to 35 T, and determined the T - and θ -dependence of B_{c2} down to 0.3 K. The anisotropy parameter $\Gamma = B_{c2}^{ab}/B_{c2}^c$ is slightly larger than typical values found in the “122” and “11” systems, but is still small, indicating quasi-isotropic superconductivity. Its temperature dependence is similar to those observed in the “122” and “11” systems. The detailed analyses show that, while B_{c2} for $B_a \parallel c$ is limited by the orbital effect, the weaker orbital effect for $B_a \parallel ab$ due to the mass anisotropy is partly compensated for by the spin-paramagnetic effect leading to a reduced anisotropy at low temperatures.

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